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Using shaped pulses to probe energy deposition during laser-induced damage of SiO_2 surfaces

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Abstract
Laser-induced damage initiation in silica has been shown to follow a power-law behavior with respect to pulse-length. Models based on thermal diffusion physics can successfully predict this scaling and the effect of pulse shape for pulses between about 3ns and 10ns. In this work we use sophisticated new measurement techniques and novel pulse shape experiments to test the limits of this scaling. We show that simple pulse length scaling fails for pulses below about 3ns. Furthermore, double pulse initiation experiments suggest that energy absorbed by the first pulse is lost on time scales much shorter than would be predicted for thermal diffusion. This time scale for energy loss can be strongly modulated by maintaining a small but non-zero intensity between the pulses. By producing damage with various pulse shapes and pulse trains it is demonstrated that the properties of any hypothetical thermal absorber become highly constrained.

Key words:SiO_2, pulse duration, laser-induced damage

I. Introduction

There are two major types of laser-induced damage which occur with single exposures to laser light. Intrinsic damage is similar to DC breakdown in that it results from high electric fields during the laser pulse ripping electrons loose. Extrinsic damage results from a feature or contaminate not native to the host material catalyzing energy absorption in one way or another. The identity of these catalyzing entities has been the subject of a great deal of thought and speculation over the years, but is still not well understood. For our purposes it is not necessary to classify the catalyzing features in particular, therefore we will simply refer to them as precursors. A few properties of the precursors are important to note, namely that they are known to reduce the electric field necessary to induced damage by one or more orders of magnitude but only act locally (typical size is believed to be a few hundred nm). The localized nature of the absorption is illustrated in Figure 1, an SEM of individual laser induced damage sites distributed semi-randomly throughout the frame.

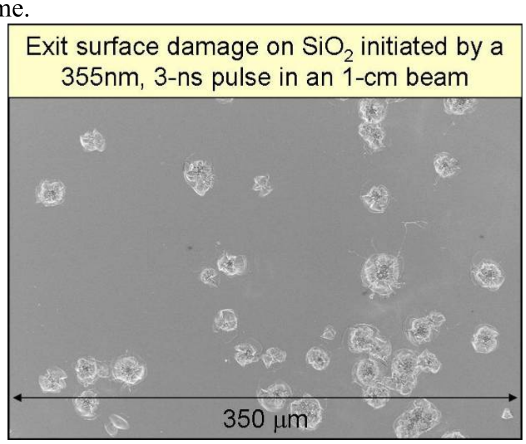

Figure 1. A SEM picture of a small fraction of the exit surface damage on SiO2 produced by a 1-cm diameter beam.

Much work has also been done in an effort to understand the effect of pulse duration and shape on damage initiation[1-7]. Most of this work has been done with laser spot sizes on the order of a few tens of microns. While such a configuration is very well suited for studying intrinsic damage behavior it is not, in general, suitable to study extrinsic damage behavior which requires the testing of much larger volumes of material[8-10].

Most previous work has cited an approximate square root dependence of initiation density on pulse duration[1,2,11,12]. Recently we have shown that a simple approximation for the heat diffusion equation can reproduce both the reported square root dependence on pulse duration as well as the observed effect of temporal pulse shape on initiation[4].

II. Results

To evaluate and expand this study of the effect of temporal pulse shape on damage initiation, with an emphasis on isolating and studying extrinsic initiations, we use the damage test techniques reported earlier[8]. In brief, a 1-cm diameter laser beam from the third harmonic (351 nm) of a Nd:Glass laser with the ability to produce arbitrary pulse shapes within a few hundred ps temporal resolution is used to induce damage on the exit surface of SiO_2 windows. Flat-In-Time (FIT) pulses are used at all pulse durations to simplify modeling the experimental results. Damage is initiated with a single pulse on test samples not previously exposed to laser radiation. A near-field image of the laser spot at an equivalent plane is captured and the individual damage sites are cataloged for size and location with an automated microscope (see Figure 2a-c).

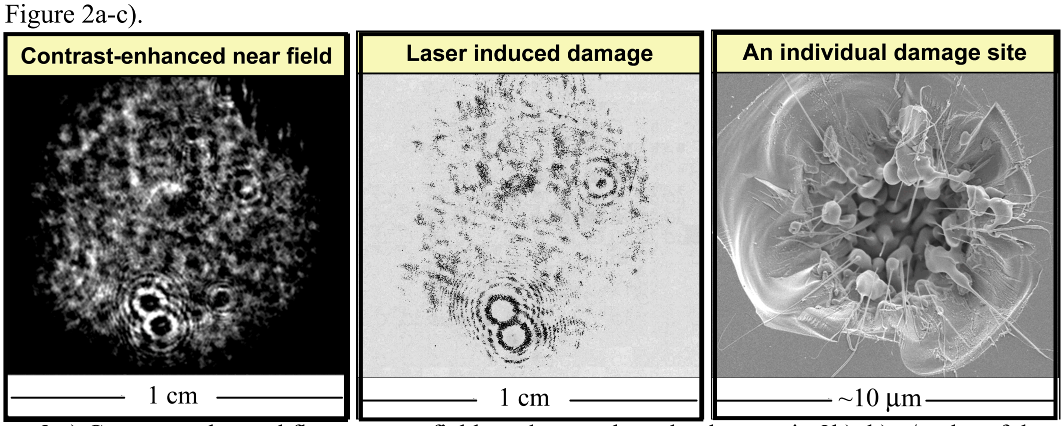

Figure 2 a) Contrast enhanced fluence near-field used to produce the damage in 2b). b) x/y plot of the damage produced. C) SEM image of an individual damage site

The damage is spatially registered to fluence near field to determine the local fluence within each damage site. This information can then be used to produce damage density vs fluence ($\rho(\phi)$) and average damage site diameter vs fluence ($d(\phi)$) curves (see Figure 3). Although this technique as described in reference 8 requires a relatively high output laser, a clever adaptation using a smaller laser has more recently been described by Lamaignere and co-workers[10].

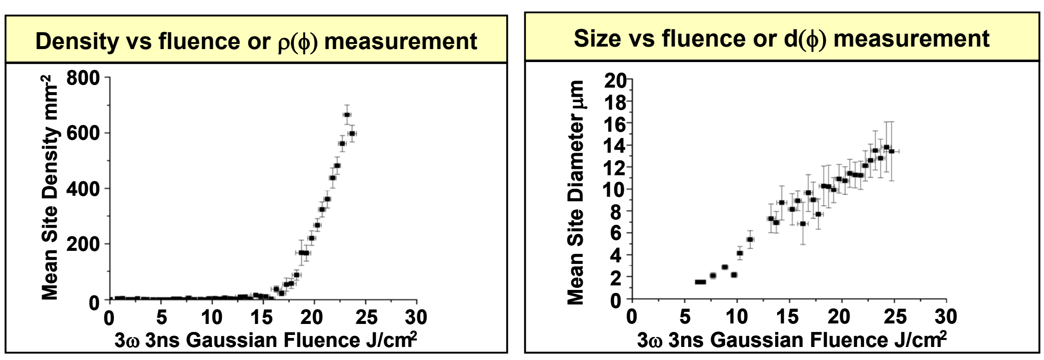

Figure 3. Typical $\rho(\phi)$ and $d(\phi)$ graphs obtained from our single shot damage test technique with a 3 ns Gaussian pulse

The $\rho(\phi)$ for a number of FIT pulses ranging from 0.35 ns to 20 ns in duration is shown in Figure 4a. We have also confirmed our previous observations that the size of damage site initiated with FIT pulses is relatively insensitive to fluence, but the size does increase approximately linearly with pulse duration (see Figure 4b).

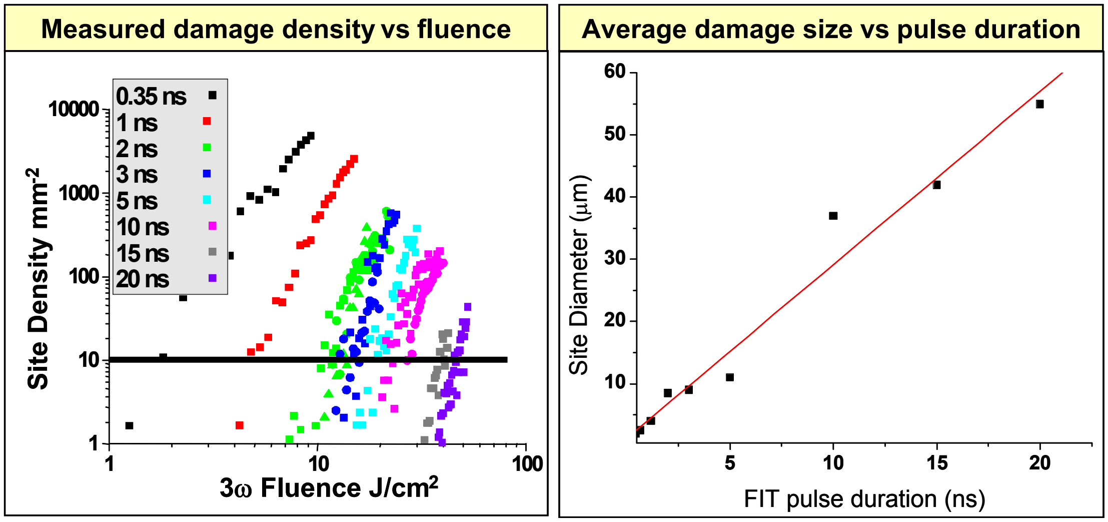

Figure 4 a) Damage density as a function of fluence for the listed pulse durations. b) Average initiation size as a function of pulse duration

The pulse length dependence of the damage density can most easily be compared to historical data by considering the level of fluence needed to produce a fixed damage density as a function of pulse duration. This treatment should resemble classical damage threshold measurements which determine the lowest fluence that will cause at least one damage site in a fixed area (defined by the spot size). Effectively a damage threshold measurement measures a damage density corresponding to one site per beam area. Typical damage threshold tests used beams of a few millimeters in diameter. For a 3 mm diameter beam with a Gaussian spatial distribution it is reasonable to assume from the power law form of the $\rho(\phi)$ in figure 2a that most if not all damage will occur in the top 10% of the beam. Thus a 3 mm diameter beam will have an effective area of ~0.1 mm^2, and a damage density of ~10 sites / mm^2 should correspond reasonably well with previously reported 'damage threshold' values. Figure 5 makes this comparison with the earlier work of Campbell & Rainer and finds reasonable agreement.

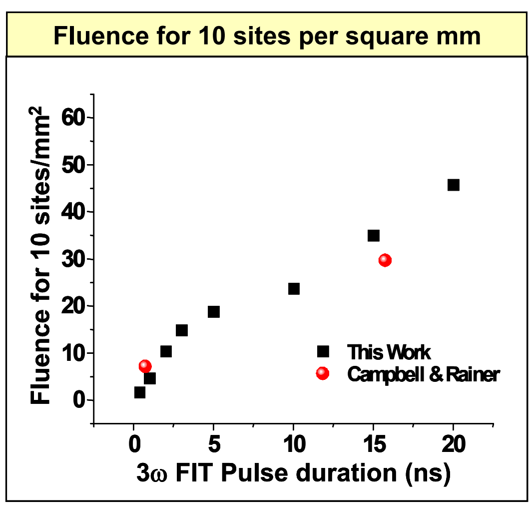

Figure 5. Comparison of this work to Campbell & Rainer damage threshold measurements. We have assumed that Campbell & Rainer had a detection limit of 10 sites per square millimeter

III. Analysis

Although the two data points from the historical data match quite well with our observations, the interpretation of a square root pulse scaling clearly does not fit our data. By recasting our $\rho(\phi)$ data as density vs intensity or $\rho(I)$ we see that the data falls into two groups, one consisting of the long pulse durations (10 ns – 20 ns) and the other containing the shorter pulses (0.35 ns – 3 ns). The data for the 5 ns pulse falls in between the two groups. The plot of intensity needed to produce a constant 10 sites per mm^2 shows that for the conditions studied here a fixed intensity of 5 GW/cm^2 will produce a constant damage density for short pulse durations. The same damage density is produced by ~2.5 GW/cm^2 for the longer pulse durations

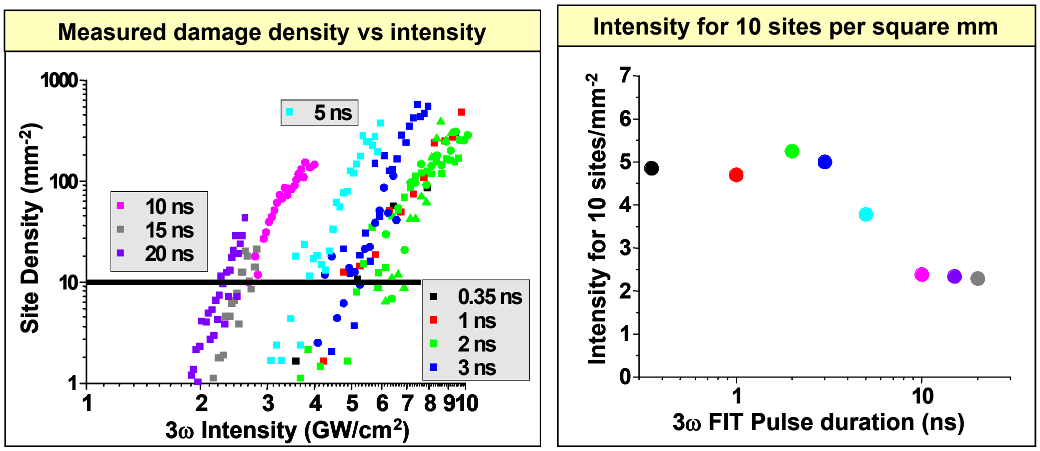

Figure 6. a) Damage density as a function of intensity. b) Intensity in GW needed to produce a damage density of 10 sites per mm^2

This change in critical intensity is an indicator that the net rate at which energy is absorbed by our precursors as a function of intensity experiences an enhancement after a few ns of laser exposure. Such a change in net absorption could be due to an increase in the absorption of the precursors or to a limitation of their ability to dissipate absorbed energy. The first effect would likely be a property of the precursor itself and therefore be localized to the precursor and manifested as a function of the time the precursor is illuminated. If however, the change in net absorption is due to a reduction in the rate at which a precursor can dissipate energy, we would expect that a regional change is occurring that would take time to propagate though the host material surrounding the precursor, but which would not necessarily require constant laser illumination. To discern between these two possibilities we initiated damage with a well controlled and characterized pair of pulses separated by a variable time delay, T_D (see Figure 7a). The resulting $\rho(\phi)$ measurements for various pulse delays can be seen in Figure 7b.

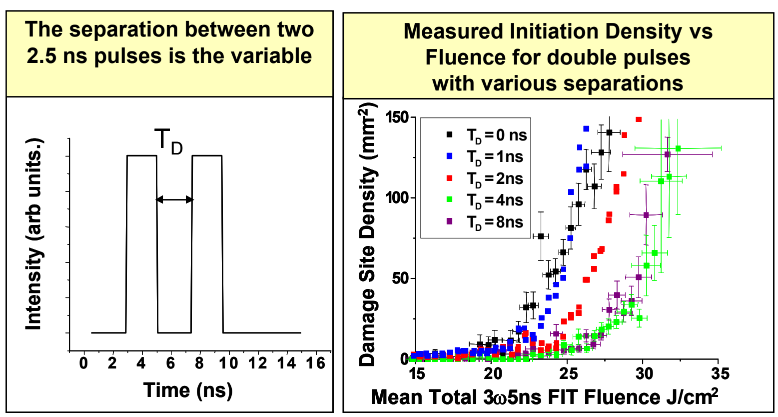

Figure. 7 a) Schematic representation of the pulse shape used in the "double pulse" experiment. b) Measured $\rho(\phi)$ for double pulses with various separation between the two pulses (T_D)

Within our experimental uncertainty our results show that for pulse separations of 0 ns, 0.5 ns (not shown), and 1 ns no statistically significant change is observed. However, once the delay is increased to 2 ns the damage density is observed to shift to higher fluence. This effect is seen to saturate with a delay of approximately 4 ns and longer. The drastic shift of damage density to higher fluence for T_D longer than

4 ns can be understood if the density of damage is plotted against the fluence contained in the first pulse only (see Figure 8).

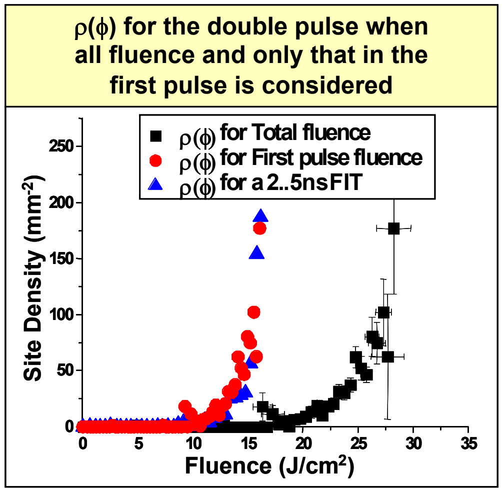

Figure 8. Measured ρ(ϕ) for a 2.5 ns FIT pulse, a double pulse with 4 ns separation, and for the double pulse with 4 ns separation when only the fluence in the first pulse is considered.

For comparison a ρ(ϕ) curve for a 2.5 ns FIT pulse from a separate experiment is included in Figure 8. The close match between the ρ(ϕ) curve for the 2.5 ns FIT pulse and that obtained by plotting the damage produced by a double pulse with a 4 ns delay vs. the fluence contained only in the first pulse indicate that the second pulse is not contributing to initiation.

Following the assumption that this result indicates a reduction in precursor's ability to dissipate energy, we postulate the cause to be neighboring precursors which are also absorbing energy and heating the host material. Such an argument would require adjacent precursors to be quite close, on the order of a few hundred nm. A lower limit to the maximum precursor density can be inferred from Figure 9 which shows a very high density of front surface damage produced under extreme conditions. It is instructive to examine front surface damage for two reasons. There is no reason to believe that precursor populations should be anything other than identical for the front and rear surface. Secondly, because damage sites initiate at such smaller sizes on the input surfaces, it is possible to observe damage as discrete sites at densities which would completely merge on the exit surface. Although not conclusive Figure 9 does suggest that a nearest neighbor spacing in the hundreds of nm is not unreasonable.

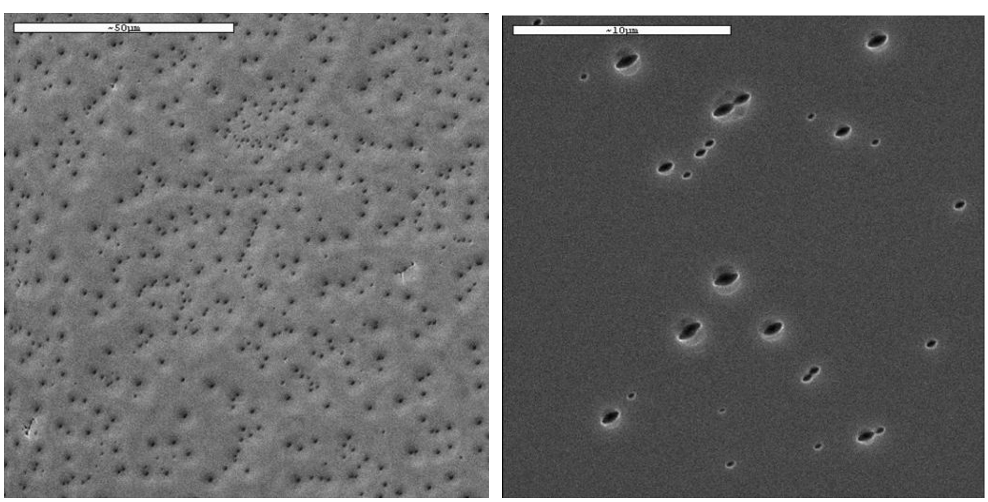

Figure 9. SEM images of ultra-high density damage produce on the front surface of a SiO_2 optic

To test the effect of this hypothesis we model the effect a lattice of absorption defects would have on the temperature surrounding a particular precursor (see Figure 10a and b). The heat equation was solved for laser heating by a flat in time 1 ns pulse of both an isolated 100 nm absorbing precursor and for a precursor with four neighbors arranged in a square. Figure 10a shows the resultant temperature histories at the central precursor.

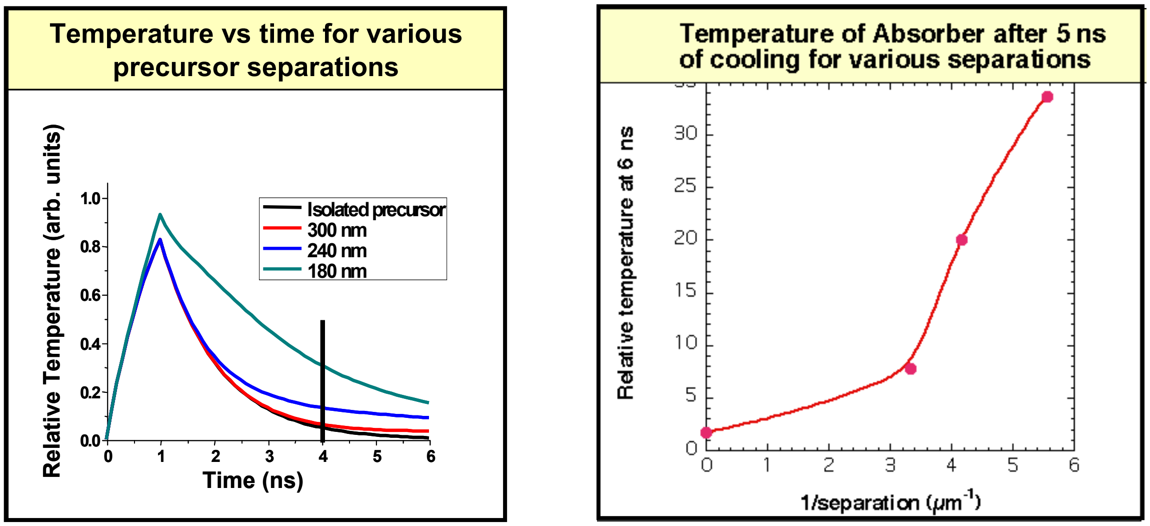

Figure 10. a) Relative temperature of a precursor as a function of time for four different nearest neighbor distances b) Relative temperature of a precursor after 6 ns as a function of nearest neighbor distance

The isolated precursor exhibits the typical temperature rise during the laser pulse followed by an exponential decay as the material cools. When there are nearby precursors, however, the cooling slows down significantly once the thermal diffusion length $(4Dt)^{1/2}$ becomes comparable to the inter-particle distance. Here D is the thermal diffusivity, $\kappa/\rho C$ where κ is the heat conductivity, ρ the density and C the heat capacity. The plot compares cooling at various interparticle distances (the isolated particle can be considered to have infinite interparticle separations). To see the implication of this, Figure 10b plots the temperature after 5 ns of cooling as a function of separation. The temperature at the shortest separation is six times higher than that of the temperature for infinite separation. This can be enough to make it easier for a subsequent pulse to reach the damage initiation temperature.

IV. Conclusion

We have reexamined the effect of pulse duration on initiation of extrinsic damage on the exit surface of SiO_2 and experimentally found results that are consistent with those previously reported by Campbell and Rainer. However, our more extensive measurements allow us to draw significantly different conclusions about the nature of pulse scaling compared to previous work. Specifically over the pulse duration range between a few hundred ps and a few tens of ns we do not see the damage density scale with the square root of the pulse duration. More over we see that for the conditions studied here the density of damage is determined by laser intensity, rather than fluence. We have argued that our results can be explained by the interaction of adjacent of precursors which interact thermally for pulses longer than a few ns. Indeed we have made several direct observations (which will be reported in the near future) of sites interacting during damage initiation.

As a parting word of caution we note that because we are studying damage initiated by extrinsic defects, all aspects of the damage response to laser parameters will depend on the precise nature of the precursors. Although it is possible and even likely that the precursors are micro-fractures left over from the polishing process, and therefore fairly universal between material, it is possible that a wide range of 'weakest' precursors are present on the surface of SiO_2 windows.

V. Acknowledgements

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